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Chemical Identification of Dubnium as a Decay Product of Element 115

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Nuclear chemistry experiments are a (huge) international team effort

Joint Institute for Nuclear Research, Dubna, Russia

Yu. Ts. Oganessian, V. K. Utyonkov, Yu. V. Lobanov, F. Sh. Abdullin, A. N. Polyakov, I. V. Shirokovsky, Yu. S. Tsyganov, G. G. Gulbekian, S. L. Bogomolov, B. N. Gikal, A. N. Mezentsev, S. Iliev, V. G. Subbotin, A. M. Sukhov, A. A. Voinov, G. V. Buklanov, K. Subotic, V. I. Zagrebaev, S. N. Dmitriev, S. V. Shishkin, A. V. Yeremin , V. I. Chepygin, E. A. Sokol, G. K. Vostokin, N. V. Aksenov, M. Hussonnois and M. G. Itkis

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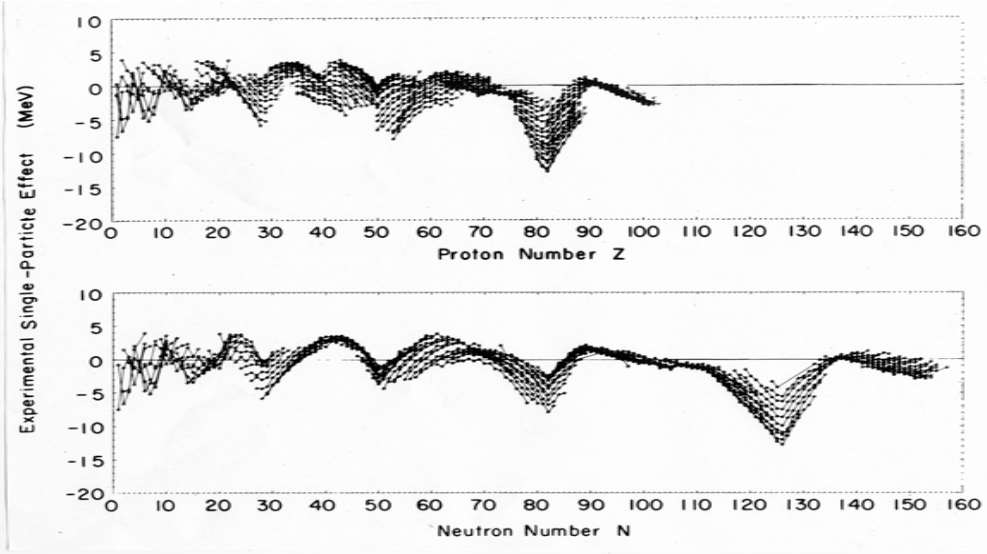
Paul Scherrer Institute, Villigen, Switzerland

D. Schumann, H. Bruchertseifer, R. Eichler, B. Eichler and H. W. Gaeggeler

The discovery of heavy elements helps to answer fundamental physics questions

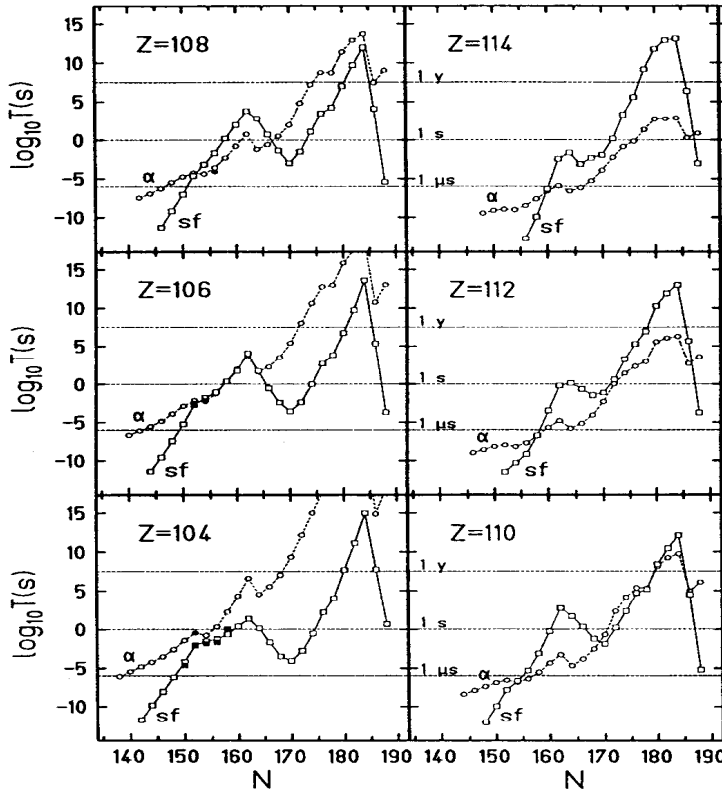
- The Liquid Drop Model successfully explains gross nuclear properties, but does not account for extra stability associated with "magic numbers" of protons or neutrons
- Magic numbers arise from second-order particle-particle interactions, resulting in shell structure in the level spacing of protons and neutrons (analogous to closed electronic shells)
- The magic numbers (so far) are 2, 8, 20, 28, 50, 82 and 126

- Magic and doubly-magic nuclei are more stable (resistant to decay) than their surrounding neighbors
- Elements with magic numbers of protons have more stable isotopes (e.g., Sn has 10)
- Doubly magic ^{132}Sn drives spontaneous fission toward asymmetric fission fragments



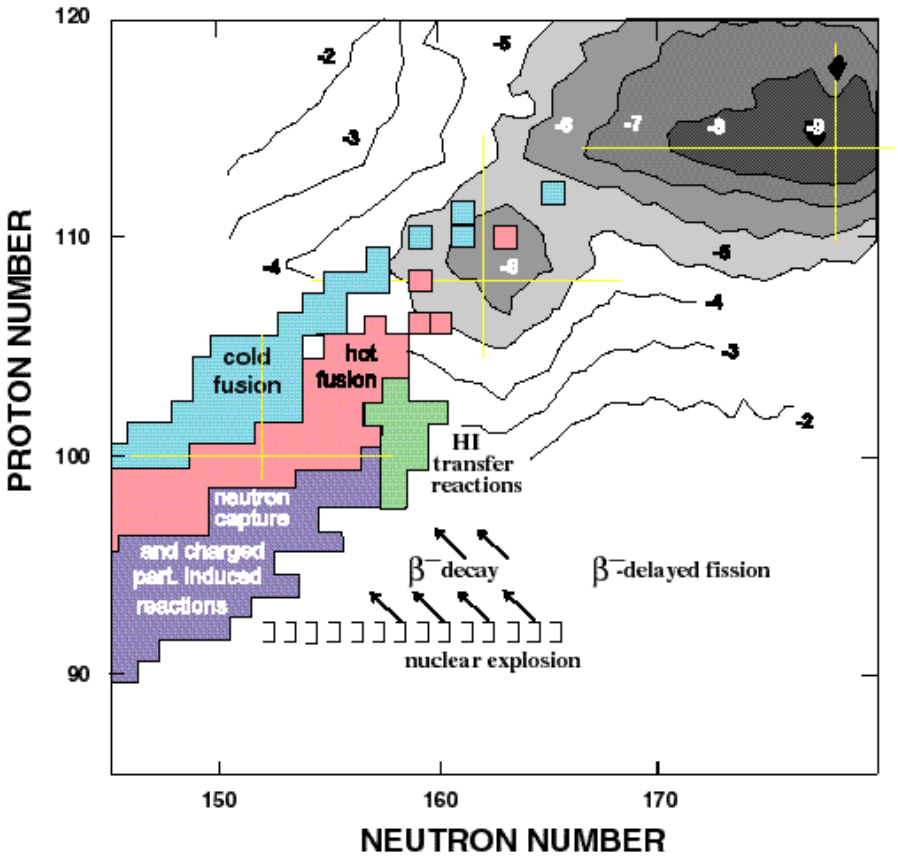
Calculating the next magic numbers remains a challenge for theorists!

Experimental data is needed to validate model calculations



With increasing nuclear charge, decay by alpha emission becomes favored over decay by spontaneous fission as one approaches the vicinity of the closed nuclear shells

The signature of the decay of a superheavy nucleus is a series of alpha decays followed by a spontaneous fission

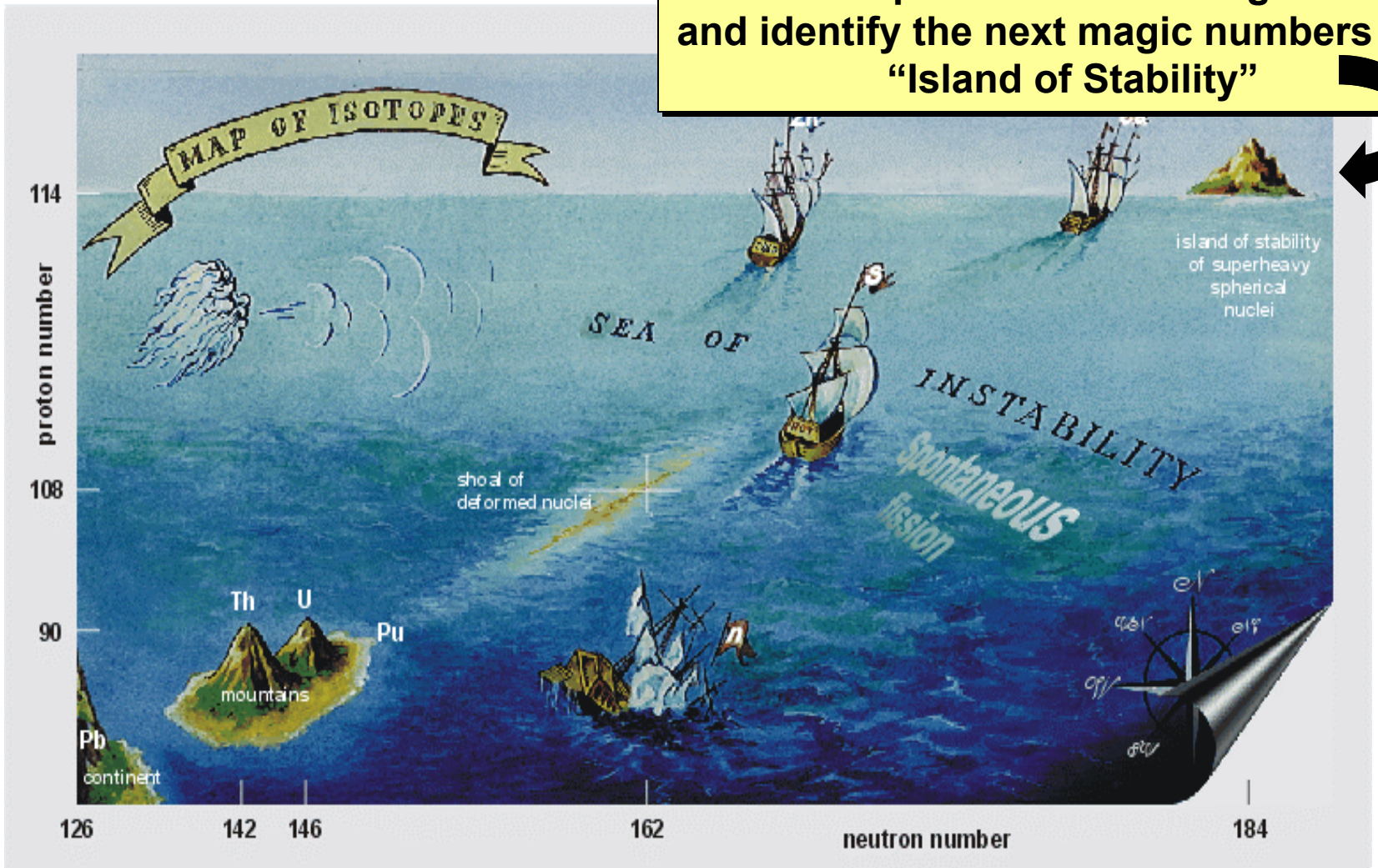


The heaviest known nuclei (circa 1998) superimposed on the calculated shell corrections to the liquid drop model

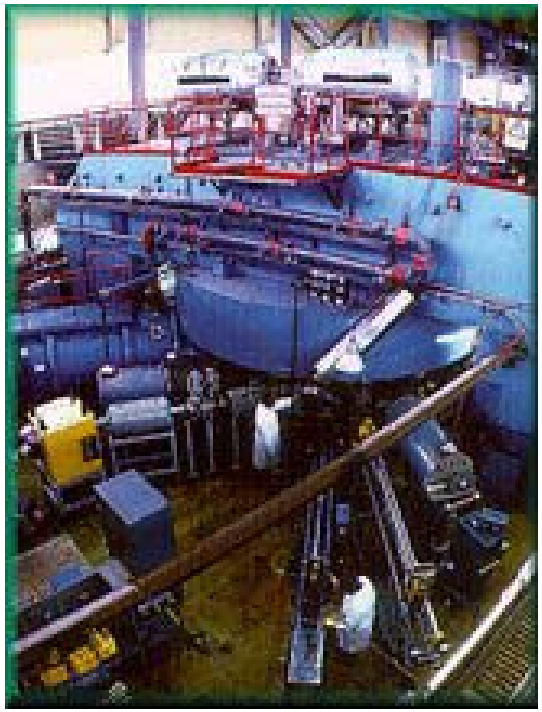
The most current theories place the next magic numbers at Z=114, N=184

Half-lives of these magic nuclei could be from seconds to days (model dependent)

Current experiments are designed to try and identify the next magic numbers - the "Island of Stability"



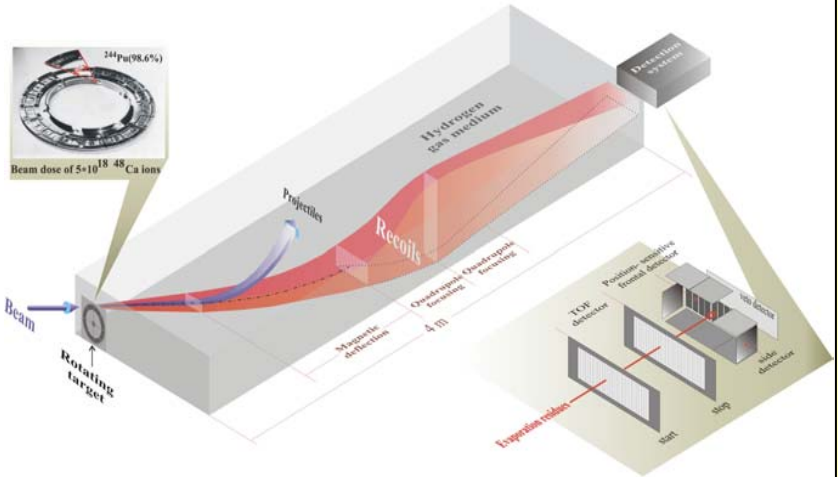
Experiments are performed at the Flerov Lab of Nuclear Reactions at JINR, Dubna, Russia



- $^{48}Ca^{5+}$ beam supplied by the U400 Cyclotron with high intensities
- Total beam dose over the course of an experiment is ~10^{18} particles

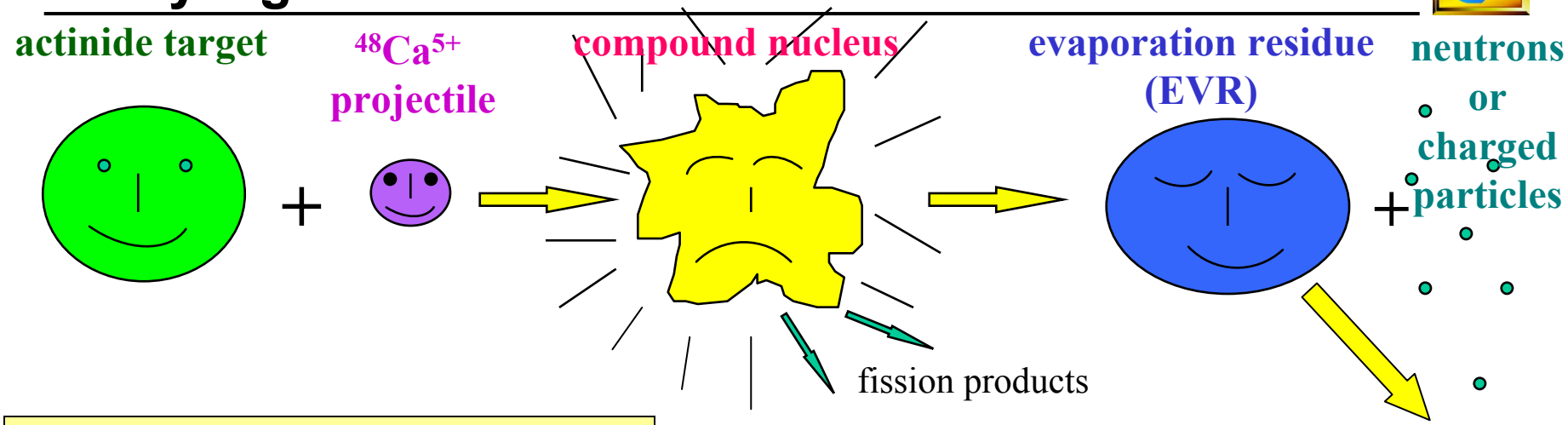
- Thin (0.35 mg/cm^2) rotating actinide oxide targets electroplated onto 1.5-μm Ti backing
- DGFRS suppression factors are $\geq 10^{15}$ and $\geq 10^4$ for beam- and target-like particles, respectively

Dubna Gas-Filled Recoil Separator (DGFRS)

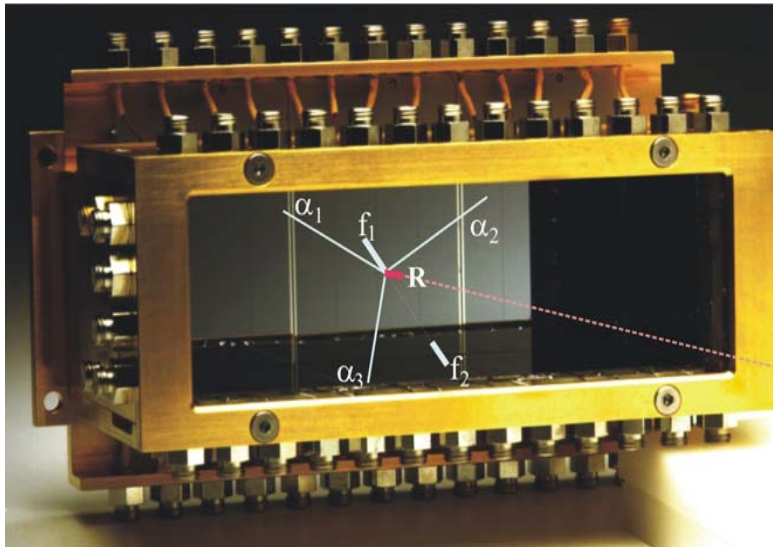


Transmission Efficiency = 35–40 % for Z = 114–116 nuclei

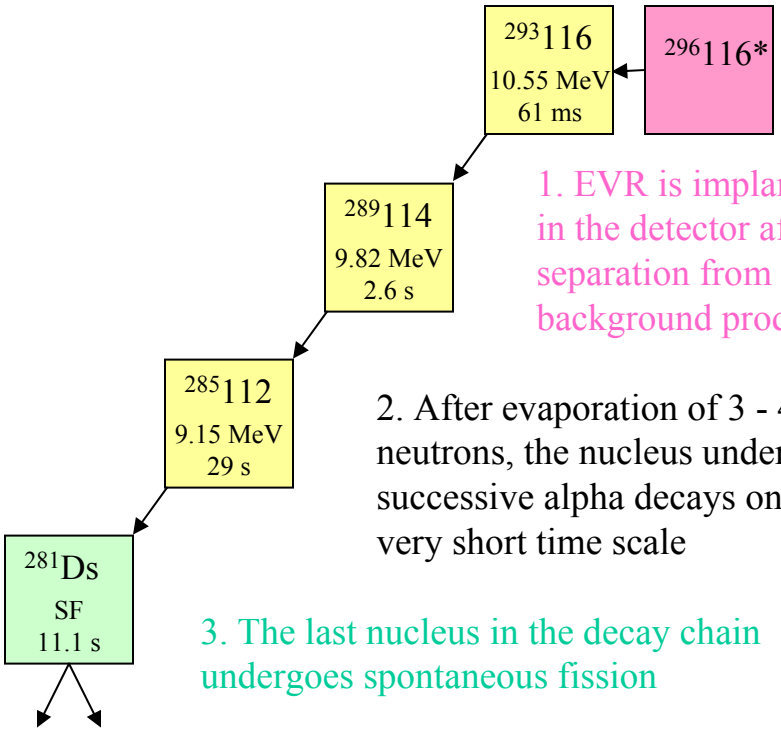
Heavy element production and detection – unique decay signatures



Position-sensitive silicon detector array



- **alpha particle efficiency = 87%**
- **coincident fission fragment efficiency = 45%**

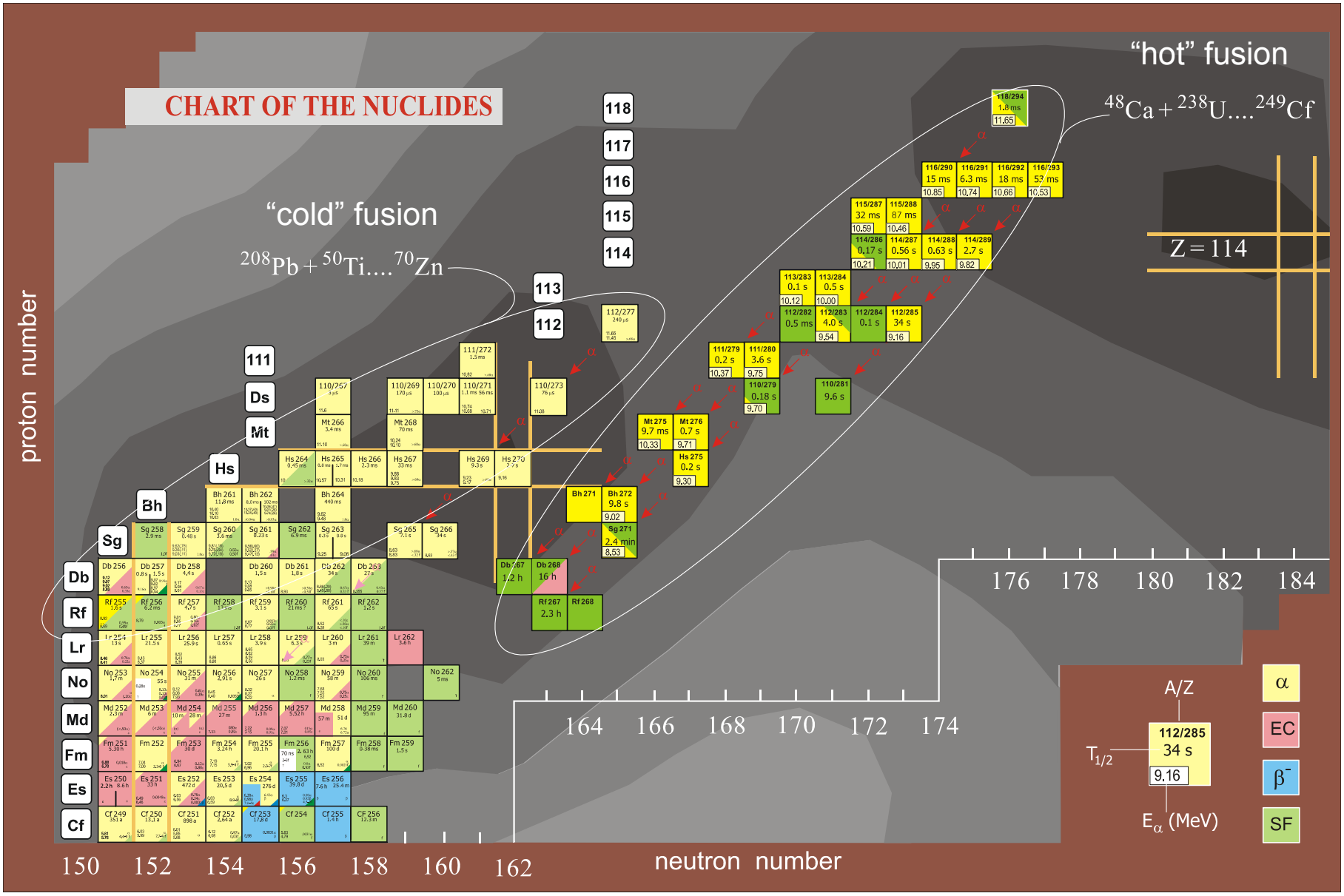


1. EVR is implanted in the detector after separation from background products

2. After evaporation of 3 - 4 neutrons, the nucleus undergoes successive alpha decays on a very short time scale

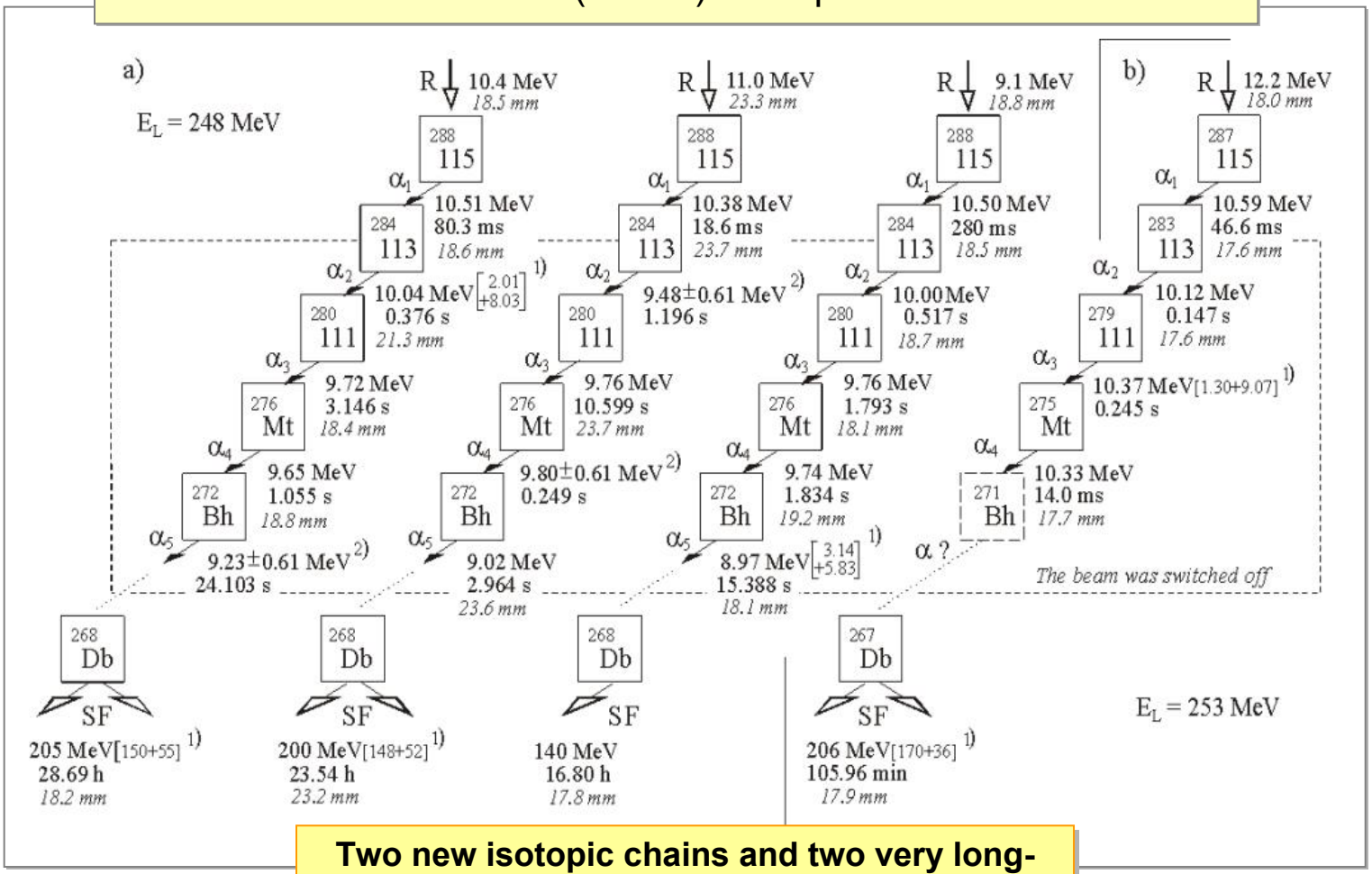
3. The last nucleus in the decay chain undergoes spontaneous fission

Chart of the nuclides – 2004



Elements 113 and 115 (2003) continue to map the neutron-deficient side of the "Island of Stability"

- $^{48}Ca + {}^{243}Am \rightarrow {}^{291}115^*$, ^{243}Am (99.9%) = 0.360 mg/cm^2
- 8.6×10^{18} ^{48}Ca ions total, beam energies = 248 and 253 MeV
- Production cross section ($^{288}115$) ≈ 2.7 pb

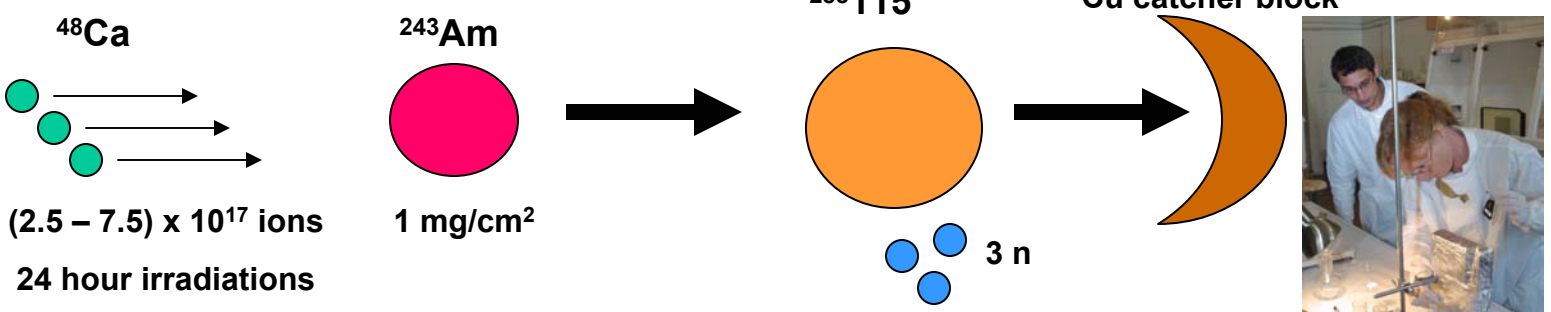


Two new isotopic chains and two very long-lived dubnium isotopes observed

The long-lived Dubnium (105) isotopes allow us to perform chemistry

- Understanding the chemical properties of the elements is the most basic problem of chemistry.
- Relativistic effects in the heavy elements dominate even in the valence orbitals
  - Simple extrapolation of periodic trends may not be valid.
  - These effects can change the shape and energy of the electronic orbitals, affecting the chemical properties of the element.
- Data is required to validate models that predict the chemical properties of the transactinides.
- Knowledge of transactinide chemical properties helps in interpreting chemical properties throughout the periodic table.
- Isolating the ^{268}Db daughter offers independent proof of the production of ^{288}115.

First attempt to identify the long-lived $^{288}115$ decay daughter – ^{268}Db ($t_{1/2} \approx 16$ hours)



- Chemistry was developed and performed by D. Schumann of PSI
- Once collected in the Cu block, $^{288}115$ undergoes five alpha decays to ^{268}Db
- The surface (10 μm) of the Cu block is shaved and dissolved in aqua regia.
- Several tracers (^{177}Ta, ^{175}Hf, ^{92m}Nb, ^{89}Zr, ^{88}Y), La carrier, and NH_4OH are added.
  - Cu remains in solution, +3, +4 and +5 ions are carried with $La(OH)_3$ precipitate
- Precipitate is washed and dissolved in HCl
- Solution is converted to nitrate form and loaded onto AG 50W-X8 cation exchange resin (nitrate form).
- Group IV and V are eluted together with 2M HF
  - +3 ions (actinides) remain on the column (no ^{88}Y detected in eluant)
- Sample is dried on thin polyethylene foil and counted
  - Solid state detectors surrounded by neutron detectors look for coincident fission fragments and number of neutrons emitted per fission
- Nb(V) yield ≈ 80%, Zr(IV) yield ≈ 60%

Results from the first ^{268}Db chemistry

	DGFRS Experiment (2003)	Chemical Experiment (2004)
Separation method	Kinematic separator	Radiochemical separation
Separation efficiency	35%	80%
Detection method	Decay chains of Z=115 nuclei	SF of Z=105 nuclei
^{48}Ca beam energy	246 MeV	247 MeV
^{48}Ca total beam dose	4.5 x 10^{18}	3.4 x 10^{18}
^{243}Am target thickness	0.36 mg/cm^2	1.2 mg/cm^2
Number of SF events	3	15
Cross section (Z=115)	2.7 (+4.8/-1.6) pb	4.2 (+1.6/-1.2) pb
Half-life (Z=105)	16 (+19/-6) h	32 (+11/-7) h
TKE of SF fragments	≈225 MeV	≈230 MeV, <v> = 4.2

But more work needs to be done:

- –This experiment proves the fission events come from the +4/+5 chemical fraction
- –The +4 and +5 ions should be separated to prove the fission events come from ^{268}Db
- –The +5 ions could also be separated from each other to show the chemical behavior of element 105 as either more Nb-like or more Ta-like

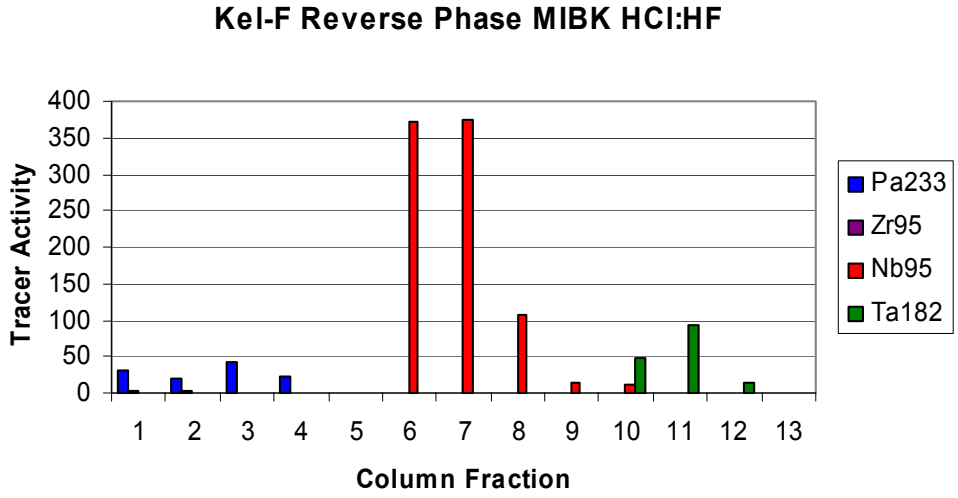
LLNL has developed a new group IV/V separation (deployed in Dubna earlier this month!)

- Step 1 – Lanthanide precipitates to remove copper from the catcher and separate +4 and +5 fractions
  - Using radiotracers of ^{233}Pa, ^{95}Zr, ^{95}Nb and ^{182}Ta
  - Add H_3BO_3 and several carriers – La^{3+} (1mg), Nb^{5+} (1μg), Ta^{5+} (1μg), and Zr^{4+} (10μg)
  - Precipitate $La(OH)_3$ with excess NH_4OH in a hot H_2O bath
    - All tracers carry down with the precipitate – Cu left in the supernatant
  - Dissolve precipitate in HCl and repeat
  - Dissolve final $La(OH)_3$ precipitate in conc. HCl
  - Add H_2O and conc. HF to form LaF_3 precipitate
  - Distribution of activities in the solid and aqueous phases:

	Pa	Zr	Nb	Ta
LaF_3 ppt.	98.5%	97.5%	11.2%	8.8%
F^- super.	1.5%	2.5%	88.8%	91.2%

Group IV/V separation continued

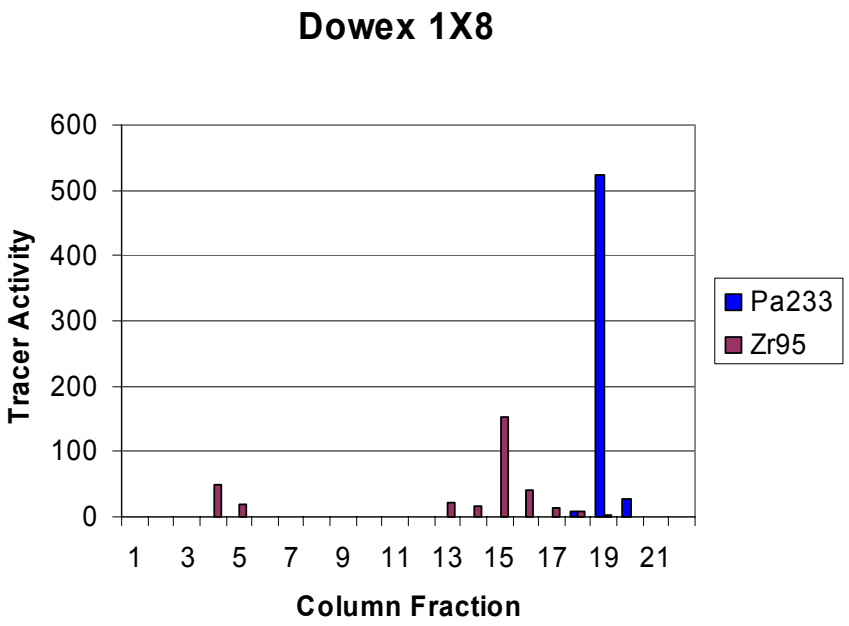
- Step 2 – Separate Nb and Ta Fractions using a reverse phase column separation
  - Supernatant from step 1 (6N HCl : 6N HF) is loaded onto a small (2mm inner diameter) Kel-F inert support column pre-conditioned with methyl isobutyl ketone (MIBK)
    - Kel-F is a fluorocarbon-based polymer similar to Teflon made from PolyChloroTriFluoroEthylene (PCTFE)
  - Nb and Ta are eluted separately using HCl:HF and water in a gradient elution



Eluant	Total Volume Added (μL)	Elements Eluted
6N HCl : 6N HF	400	Any residual Pa, Zr not removed by LaF_3 ppt.
3N HCl : 1N HF	200	Any remaining Pa
1.5N HCl : 0.5N HF	300	Nb
H_2O	1400	Ta

Group IV/V separation continued

- Step 3 – Separate Zr and Pa fractions from the LaF_3 precipitate formed in step 1
  - Dissolve LaF_3 in boric acid and conc. HCl
  - Add Sc carrier and NH_4OH to form $La(OH)_3$ precipitate
  - Dissolve precipitate in conc. HCl and load onto a 2mm i.d. Dowex 1x8 anion resin column
  - Remove Sc, La, actinides and RE contaminants with conc. HCl
  - Elute Zr and Pa using a gradient elution



Eluant	Total Volume Added (μL)	Elements Eluted
12N HCl	1900	Sc, La, actinides, rare earths, minor Zr (~ 20%)
5N HCl	2500	Zr
2N HCl	5500	Pa

Summary and future work

- The first attempt to identify the long-lived dubnium daughter of element 115 was successful although some questions remained
- New chemistry was developed at LLNL to separate the Group IV and V fractions and to isolate the individual Group V elements
  - Learn something about the chemical properties of element 105 in this system – does it behave more like Nb, Ta or Pa?
- Chemistry of element 105 was repeated this month in Dubna
- In addition to element 105 chemistry, we are currently working on an automated chemistry apparatus for element 114 chemistry experiments

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